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(19) (CA) **APPLICATION FOR CANADIAN PATENT** (12)

(54) N-Aryl-Nitrogen Heterocycles

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Notice: This application is as filed and may therefore contain an  
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**Canada**

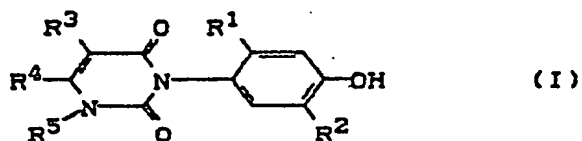
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N-Aryl-nitrogen heterocycles

## A b s t r a c t

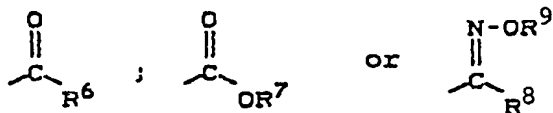
The invention relates to new N-aryl-nitrogen heterocycles of the general formula (I)



in which

R<sup>1</sup> represents hydrogen or halogen,

R<sup>2</sup> represents one of the radicals



R<sup>3</sup> represents hydrogen and

R<sup>4</sup> represents alkyl or halogenoalkyl, or

R<sup>3</sup> and R<sup>4</sup> together represent a divalent alkanediyl radical and

R<sup>5</sup> represents an in each case straight-chain or branched radical from the series comprising alkyl, alkenyl, alkynyl, halogenoalkyl or cycloalkyl,

where

R<sup>6</sup> represents hydrogen or alkyl,

R<sup>7</sup> represents an in each case optionally substituted radical from the series comprising alkyl, alkenyl or cycloalkyl,

R<sup>8</sup> represents hydrogen or alkyl and

R<sup>9</sup> represents an in each case optionally substituted radical from the series comprising alkyl, alkenyl or cycloalkyl.

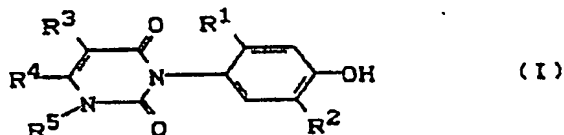
several processes for their preparation, and their use as herbicides.

The invention relates to new N-aryl-nitrogen heterocycles, to a plurality of processes for their preparation, and to their use as herbicides.

5 It is known that certain N-aryl-nitrogen heterocycles such as, for example, the compound 1-(4-cyano-2,5-difluorophenyl)-3-(1-methylethylidene)-pyrrolidine-2,5-dione, have herbicidal properties (cf., for example, DE 3,835,168). Further N-aryl-nitrogen heterocycles are described in DE 3,643,748, JP 1,052,755, GB 2,071,100 and  
10 DE 3,839,480.

However, the herbicidal activity of these previously known compounds against problem weeds as well as their tolerance by important crop plants is not entirely satisfactory in all fields of application.

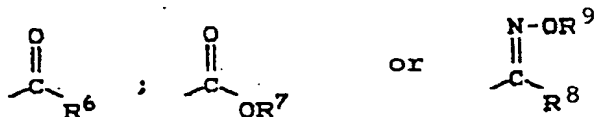
15 New N-aryl-nitrogen heterocycles of the general formula (I)



in which

R<sup>1</sup> represents hydrogen or halogen,

$R^2$  represents one of the radicals



$R^3$  represents hydrogen and

$R^4$  represents alkyl or halogenoalkyl, or

5  $R^3$  and  $R^4$  together represent a divalent alkanediyl radical and

$R^5$  represents an in each case straight-chain or branched radical from the series comprising alkyl, alkenyl, alkynyl, halogenoalkyl or cycloalkyl,

10 where

$R^6$  represents hydrogen or alkyl,

$R^7$  represents an in each case optionally substituted radical from the series comprising alkyl, alkenyl or cycloalkyl,

15  $R^8$  represents hydrogen or alkyl and

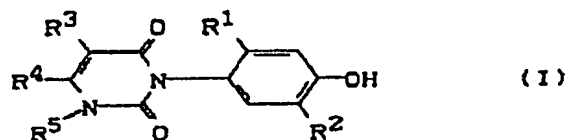
$R^9$  represents an in each case optionally substituted radical from the series comprising alkyl, alkenyl or cycloalkyl,

have now been found.

Depending on the nature of the substituents, the compounds of the formula (I) may exist in the form of geometric and/or optical isomers or isomer mixtures of

various compositions. The invention claims the pure isomers as well as the isomer mixtures.

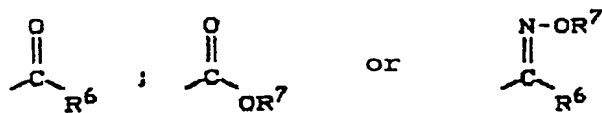
Furthermore, it has been found that the new N-aryl-nitrogen heterocycles of the general formula (I)



5 in which

R<sup>1</sup> represents hydrogen or halogen,

R<sup>2</sup> represents one of the radicals



R<sup>3</sup> represents hydrogen and

10 R<sup>4</sup> represents alkyl or halogenoalkyl, or

R<sup>3</sup> and R<sup>4</sup> together represent a divalent alkanediyl radical and

R<sup>5</sup> represents an in each case straight-chain or

branched radical from the series comprising alkyl, alkenyl, alkynyl, halogenoalkyl or cycloalkyl,

where

$R^6$  represents hydrogen or alkyl,

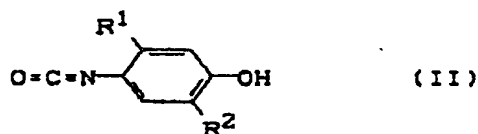
5  $R^7$  represents an in each case optionally substituted radical from the series comprising alkyl, alkenyl or cycloalkyl,

$R^8$  represents hydrogen or alkyl and

$R^9$  represents an in each case optionally substituted radical from the series comprising alkyl, alkenyl or cycloalkyl,

are obtained when

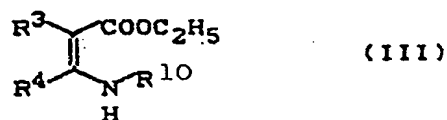
10 a) isocyanates of the formula (II)



in which

$R^1$  and  $R^2$  have the abovementioned meanings,

are reacted with  $\beta$ -enamino esters of the formula (III)



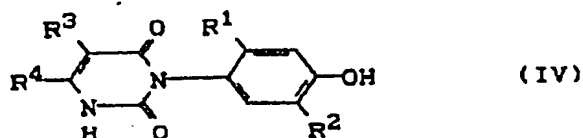
in which

$R^3$  and  $R^4$  have the abovementioned meanings and

$R^{10}$  represents hydrogen or an in each case straight-chain or branched radical from the series comprising alkyl, alkenyl, alkynyl, halogenoalkyl or cycloalkyl,

if appropriate in the presence of a diluent and if appropriate in the presence of a reaction auxiliary, and, if desired,

b) the resulting N-aryl-nitrogen heterocycles of the formula (IV)



in which

$R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  have the abovementioned meanings

are subsequently reacted with alkylating agents of the formula (V)



in which

$R^5$  has the abovementioned meaning and

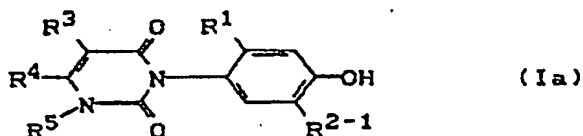


E represents an electron-attracting leaving group,

if appropriate in the presence of a diluent and if appropriate in the presence of a reaction auxiliary, or when

5

- c) the N-aryl-nitrogen heterocycles which can be obtained with the aid of process (a) or (b) according to the invention, of the formula (Ia)



in which

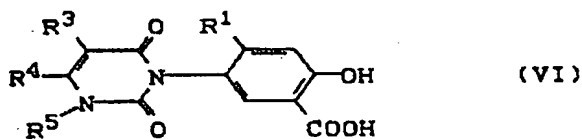
10

R<sup>1</sup>, R<sup>3</sup>, R<sup>4</sup> and R<sup>5</sup> have the abovementioned meanings and

R<sup>2-1</sup> represents an alkoxycarbonyl radical,

are first hydrolysed with acid on the ester group in the aryl moiety, if appropriate in the presence of a diluent, and the resulting N-aryl-nitrogen heterocycles of the formula (VI)

15



in which

$R^1$ ,  $R^3$ ,  $R^4$  and  $R^5$  have the abovementioned meanings, are esterified in a subsequent 2nd step with alcohols of the formula (VII)



5 in which

$R^7$  has the abovementioned meaning,

if appropriate in the presence of a diluent and if appropriate in the presence of a reaction auxiliary.

10 Finally, it has been found that the new N-aryl-nitrogen heterocycles of the general formula (I) have herbicidal properties.

15 Surprisingly, the N-aryl-nitrogen heterocycles of the general formula (I) according to the invention show a considerably better herbicidal activity against problem weeds combined with an equally good compatibility with important crop plants compared with the N-aryl-nitrogen heterocycles known from the prior art such as, for example, the compound 1-(4-cyano-2,5-difluorophenyl)-3-(1-methylethylidene)-pyrrolidine-2,5-dione, which are  
20 similar compounds chemically and from the point of view of their action.

Formula (I) provides a general definition of the N-aryl-

nitrogen heterocycles according to the invention.  
Preferred compounds of the formula (I) are those in which

$R^1$  represents hydrogen, fluorine, chlorine, bromine or iodine,

5  $R^2$  represents one of the radicals



$R^3$  represents hydrogen and

10  $R^4$  represents in each case straight-chain or branched alkyl having 1 to 6 carbon atoms or halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, or

$R^3$  and  $R^4$  together represent a divalent straight-chain or branched alkanediyl radical having 3 to 10 carbon atoms, and

15  $R^5$  represents in each case straight-chain or branched radical from the series comprising alkyl having 1 to 6 carbon atoms, halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, alkenyl having 2 to 6 carbon atoms,

alkynyl having 2 to 6 carbon atoms, or cycloalkyl having 3 to 8 carbon atoms,

where

5         $R^6$     represents hydrogen or straight-chain or branched alkyl having 1 to 6 carbon atoms

10         $R^7$     represents an in each case straight-chain or branched radical from the series comprising alkyl having 1 to 12 carbon atoms, halogeno-alkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, alkoxy-alkyl, alkoxyalkoxyalkyl or alkoxycarbonyl-alkyl, each of which has 1 to 6 carbon atoms in the individual alkyl moieties, alkenyl having 2 to 6 carbon atoms, or cycloalkyl which has 3 to 8 carbon atoms and which is optionally monosubstituted or polysubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 4 carbon atoms,

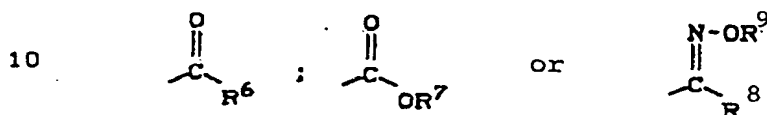
15         $R^8$     represents hydrogen or straight-chain or branched alkyl having 1 to 6 carbon atoms and

20         $R^9$     represents an in each case straight-chain or branched radical from the series comprising alkyl having 1 to 12 carbon atoms, halogeno-alkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, alkoxy-alkyl, alkoxyalkoxyalkyl or alkoxycarbonyl-alkyl, each of which has 1 to 6 carbon atoms in the individual alkyl moieties, alkenyl having 2 to 6 carbon atoms, or cycloalkyl which has 3 to 8 carbon atoms and which is optionally monosubstituted or polysubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 4 carbon atoms.

Particularly preferred compounds of the formula (1) are those in which

R<sup>1</sup> represents hydrogen, fluorine, chlorine or bromine,

R<sup>2</sup> represents one of the radicals



15 R<sup>3</sup> represents hydrogen and

R<sup>4</sup> represents in each case straight-chain or branched alkyl having 1 to 4 carbon atoms or halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, or

R<sup>3</sup> and R<sup>4</sup> together represent a divalent straight-chain or branched alkanediyl radical having 3 to 8 carbon atoms, and

25 R<sup>5</sup> represents in each case straight-chain or branched radical from the series comprising alkyl having 1 to 4 carbon atoms, halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, alkenyl having 2 to 5 carbon atoms, alkynyl having 2 to 5 carbon atoms, or cycloalkyl having 3 to 7 carbon atoms,

where

35 R<sup>6</sup> represents hydrogen or straight-chain or branched alkyl having 1 to 3 carbon atoms,

- 5 R<sup>7</sup> represents an in each case straight-chain or  
branched radical from the series comprising  
alkyl having 1 to 8 carbon atoms, halogenoalkyl  
having 1 to 4 carbon atoms and 1 to 9 identical  
or different halogen atoms, alkoxyalkyl,  
10 alkoxyalkoxyalkyl or alkoxycarbonylalkyl, each  
of which has 1 to 4 carbon atoms in the indivi-  
dual alkyl moieties, alkenyl having 2 to 4  
carbon atoms, or cycloalkyl which has 3 to 7  
carbon atoms and is optionally monosubstituted  
to trisubstituted by identical or different  
straight-chain or branched alkyl substituents  
having 1 to 3 carbon atoms,
- 15 R<sup>8</sup> represents hydrogen or straight-chain or  
branched alkyl having 1 to 3 carbon atoms and
- R<sup>9</sup> represents an in each case straight-chain or  
branched radical from the series comprising  
alkyl having 1 to 8 carbon atoms, halogenoalkyl  
having 1 to 4 carbon atoms and 1 to 9 identical  
or different halogen atoms, alkoxyalkyl,  
alkoxyalkoxyalkyl or alkoxycarbonylalkyl, each  
of which has 1 to 4 carbon atoms in the indivi-  
dual alkyl moieties, alkenyl having 2 to 4  
carbon atoms, or cycloalkyl which has 3 to 7  
carbon atoms and is optionally monosubstituted  
to trisubstituted by identical or different  
straight-chain or branched alkyl substituents  
having 1 to 3 carbon atoms.

Very particularly preferred compounds of the formula (I)  
are those in which

$R^1$  represents hydrogen or fluorine,

$R^2$  represents one of the radicals

10



15

$R^3$  represents hydrogen and

20

$R^4$  represents in each case straight-chain or branched  
alkyl having 1 to 3 carbon atoms or halogenoalkyl  
having 1 to 2 carbon atoms and 1 to 5 identical or  
different halogen atoms, or

25

$R^3$  and  $R^4$  together represent a divalent straight-  
chain or branched alkanediyl radical  
having 3 to 6 carbon atoms, and

30

$R^5$  represents an in each case straight-chain or  
branched radical from the series comprising alkyl  
having 1 to 3 carbon atoms, halogenoalkyl having 1  
to 2 carbon atoms and 1 to 5 identical or different  
halogen atoms, alkenyl having 2 to 4 carbon atoms,  
alkinyl having 2 to 4 carbon atoms, or cyclohexyl,

35

where

R<sup>6</sup> represents hydrogen or straight-chain or branched alkyl having 1 to 3 carbon atoms, and

5 R<sup>7</sup> represents an in each case straight-chain or branched radical from the series comprising alkyl having 1 to 6 carbon atoms, halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, alkoxyalkyl, alkoxyalkoxyalkyl or alkoxycarbonylalkyl, each of which has 1 to 3 carbon atoms in the individual alkyl moieties, alkenyl having 2 to 3 carbon atoms, or represents cyclopropyl, cyclopentyl or cyclohexyl, each of which is optionally monosubstituted or disubstituted by methyl,

10

15

20 R<sup>8</sup> represents hydrogen or straight-chain or branched alkyl having 1 to 3 carbon atoms, and

R<sup>9</sup> represents an in each case straight-chain or branched radical from the series comprising alkyl having 1 to 6 carbon atoms, halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, alkoxyalkyl, alkoxyalkoxyalkyl or alkoxycarbonylalkyl, each of which has 1 to 3 carbon atoms in the individual alkyl moieties, alkenyl having 2 to 3 carbon atoms, or represents cyclopropyl, cyclopentyl or cyclohexyl, each of which is optionally monosubstituted or disubstituted by methyl.

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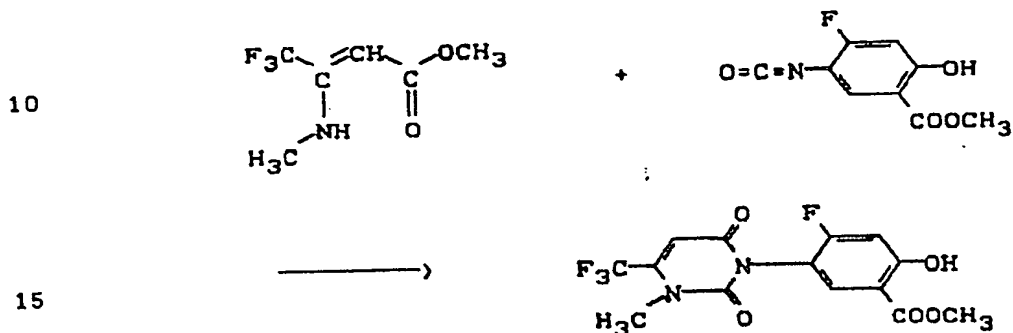
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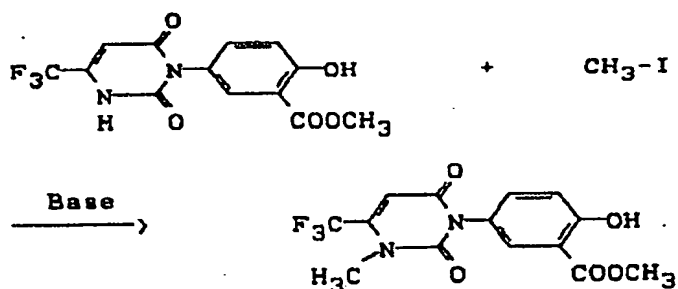
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If, for example, 2-fluoro-4-hydroxy-5-methoxycarbonyl-phenyl isocyanate and methyl 3-(N-methylamino)-4,4,4-trifluoro-crotonate are used as starting materials, the course of the reaction of process (a) according to the invention can be represented by the following equation:

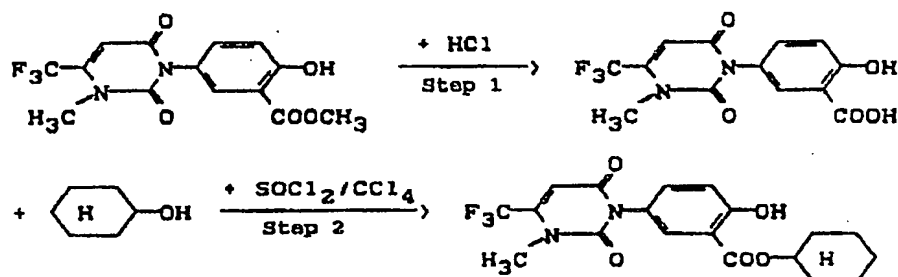


If, for example, 3-(4-hydroxy-3-methoxycarbonyl-phenyl)-6-trifluoromethyl-1,2,3,4-tetrahydro-1H-pyrimidine-2,4-dione and methyl iodide are used as starting materials, the course of the reaction of process (b) according to the invention can be represented by the following equation:

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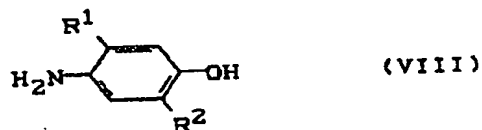
5 If, for example, 3-(4-hydroxy-3-methoxycarbonyl-phenyl)-1-methyl-6-trifluoromethyl-1,2,3,4-tetrahydro-pyrimidine-2,4-dione and cyclohexanol are used as starting materials, the course of the reaction of process (c) according to the invention can be represented by the following equation:



10 Formula (II) provides a general definition of the isocyanates required as starting materials for carrying out process (a) according to the invention. In this formula (II),  $R^1$  and  $R^2$  preferably represent those radicals which have already been mentioned in connection with the

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description of the substances of the formula (I) according to the invention as being preferred for these substituents. The isocyanates of the formula (II) were hitherto unknown and are also a subject of the invention. They are obtained when aniline derivatives of the formula (VIII)



in which

R<sup>1</sup> and R<sup>2</sup> have the abovementioned meaning,

are reacted with trichloromethyl chloroformate (diphosgene) at temperatures between 50°C and 150°C, if appropriate in the presence of a diluent such as, for example, ethyl acetate.

Aniline derivatives of the formula (VIII) are known or can be obtained in analogy to known processes (cf., for example, EP 291,159; EP 275,221; EP 253,788; EP 262,618; DE 3,638,364; PCT WO 8603194; J. Org. Chem. 54, 3740-3744 [1989]).

Formula (III) provides a general definition of the β-enamino esters required as starting materials for carrying out process (a) according to the invention. In this formula (III), R<sup>3</sup> and R<sup>4</sup> preferably represent those

radicals which have already been mentioned in connection with the description of the substances of the formula (I) according to the invention as being preferred for these substituents.

5  $R^{10}$  preferably represents hydrogen or an in each case straight-chain or branched radical from the series comprising alkyl having 1 to 6 carbon atoms, halogeno-alkyl having 1 to 6 carbon atoms and 1 to 13 identical or  
10 different halogen atoms, alkenyl having 2 to 6 carbon atoms, alkynyl having 2 to 6 carbon atoms, or cycloalkyl having 3 to 8 carbon atoms.

$R^{10}$  particularly preferably represents an in each case straight-chain or branched radical from the series comprising alkyl having 1 to 4 carbon atoms, halogeno-alkyl having 1 to 4 carbon atoms and 1 to 9 identical or  
15 different halogen atoms, alkenyl having 2 to 5 carbon atoms, alkynyl having 2 to 5 carbon atoms, or cycloalkyl having 3 to 7 carbon atoms.

The  $\beta$ -enamino esters of the formula (III) are known or  
20 can be obtained in analogy to known processes (cf., for example, PCT WO 9107392; EP 304,409; J. Heterocycl. Chem. 9, 513-522 [1972]; EP 295,233).

Formula (IV) provides a general definition of the N-aryl-nitrogen heterocycles required as starting materials for  
25 carrying out process (b) according to the invention. In this formula (IV),  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  preferably represent

those radicals which have already been mentioned in connection with the description of the substances of the formula (I) according to the invention as being preferred for these substituents.

5 The N-aryl-nitrogen heterocycles of the formula (IV) were hitherto unknown and are also a subject of the invention. They are obtained with the aid of process (a) according to the invention.

10 Formula (V) provides a general definition of the alkylating agents furthermore required as starting materials for carrying out process (b) according to the invention. In this formula (V),  $R^5$  preferably represents those radicals which have already been mentioned in connection with the description of the substances of the  
15 formula (I) according to the invention as being preferred for these substituents.

E represents a leaving radical customary in alkylating agents, preferably halogen, in particular chlorine, bromine or iodine, or in each case optionally substituted  
20 alkylsulphonyloxy, alkoxysulphonyloxy or arylsulphonyloxy such as, in particular, methanesulphonyloxy, trifluoromethanesulphonyloxy, methoxysulphonyloxy, ethoxysulphonyloxy or p-toluenesulphonyloxy.

25 The alkylating agents of the formula (V) are generally known compounds of organic chemistry.

Formula (Ia) provides a general definition of the N-aryl-nitrogen heterocycles required as starting materials for carrying out process (c) according to the invention. In this formula (Ia),  $R^1$ ,  $R^3$ ,  $R^4$  and  $R^5$  preferably represent those radicals which have already been mentioned in connection with the description of the substances of the formula (I) according to the invention as being preferred for these substituents.

$R^{2-1}$  preferably represents a straight-chain or branched alkoxy carbonyl radical having 1 to 4 carbon atoms in the alkoxy moiety, in particular a methoxy carbonyl radical or an ethoxy carbonyl radical.

The N-aryl-nitrogen heterocycles of the formula (Ia) are compounds according to the invention and can be obtained with the aid of processes (a) and/or (b) according to the invention.

Formula (VI) provides a general definition of the N-aryl-nitrogen heterocycles which occur as intermediates when carrying out process (c) according to the invention. In this formula (VI),  $R^1$ ,  $R^3$ ,  $R^4$  and  $R^5$  preferably represent those radicals which have already been mentioned in connection with the description of the substances of the formula (I) according to the invention as being preferred for these substituents.

The N-aryl-nitrogen heterocycles of the formula (VI) were hitherto unknown and are also a subject of the invention. They are obtained with the aid of process (c) according to the invention.

Formula (VII) provides a general definition of the alcohols furthermore required as starting materials for carrying out process (c) according to the invention. In this formula (VII), R<sup>7</sup> preferably represents those radicals which have already been mentioned in connection with the description of the substances of the formula (I) according to the invention as being preferred for these substituents.

The alcohols of the formula (VII) are generally known compounds of organic chemistry.

Suitable diluents for carrying out process (a) according to the invention are inert organic solvents. These include, in particular, aliphatic, alicyclic or aromatic, optionally halogenated hydrocarbons such as, for example, benzene, toluene, xylene, chlorobenzene, dichlorobenzene, petroleum ether, hexane, cyclohexane, dichloromethane, chloroform or carbon tetrachloride; ethers such as diethyl ether, diisopropyl ether, dioxane, tetrahydrofuran or ethylene glycol dimethyl ether or ethylene glycol diethyl ether; ketones such as acetone or butanone or methyl isobutyl ketone; nitriles such as acetonitrile, propionitrile or benzonitrile; amides such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide, esters such as methyl acetate or ethyl acetate, or sulphoxides such as dimethyl sulphoxide.

Process (a) according to the invention is preferably

carried out in the presence of a suitable reaction auxiliary. Suitable reaction auxiliaries are all customary inorganic or organic bases. These include, for example, alkaline earth metal hydroxides or alkali metal hydroxides such as sodium hydroxide, calcium hydroxide, potassium hydroxide or else ammonium hydroxide, alkali metal carbonates such as sodium carbonate, potassium carbonate, potassium hydrogen carbonate, sodium hydrogen carbonate or ammonium carbonate, alkali metal acetates or alkaline earth metal acetates such as sodium acetate, potassium acetate, calcium acetate or ammonium acetate, and also tertiary amines such as trimethylamine, triethylamine, tributylamine, N,N-dimethylaniline, pyridine, piperidine, N-methylpiperidine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

When carrying out process (a) according to the invention, the reaction temperatures can be varied within a substantial range. In general, the process is carried out at temperatures between  $-70^{\circ}\text{C}$  and  $+50^{\circ}\text{C}$ , preferably at temperatures between  $-40^{\circ}\text{C}$  and  $+20^{\circ}\text{C}$ .

For carrying out process (a) according to the invention, 1.0 to 1.5 moles, preferably 1.0 to 1.2 moles, of amino ester of the formula (III) and, if appropriate, 1.0 to 5.0 moles, preferably 1.0 to 2.5 moles, of the base are generally employed as reaction auxiliary per mole of isocyanate of the formula (II).

The reaction is carried out and the reaction products are



worked up and isolated by generally customary methods (cf. in this context the preparation examples).

Suitable diluents for carrying out process (b) according to the invention are inert organic solvents. These include, in particular, aliphatic, alicyclic or aromatic, optionally halogenated hydrocarbons such as, for example, benzene, toluene, xylene, chlorobenzene, dichlorobenzene, petroleum ether, hexane, cyclohexane, dichloromethane, chloroform or carbon tetrachloride; ethers such as diethyl ether, diisopropyl ether, dioxane, tetrahydrofuran or ethylene glycol dimethyl ether or ethylene glycol diethyl ether; ketones such as acetone or butanone or methyl isobutyl ketone; nitriles such as acetonitrile, propionitrile or benzonitrile; amides such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide, esters such as methyl acetate or ethyl acetate, or sulphoxides such as dimethyl sulphoxide.

If appropriate, process (b) according to the invention can also be carried out in a two-phase system such as, for example, water/toluene or water/dichloromethane, if appropriate in the presence of a phase transfer catalyst. Examples of such catalysts which may be mentioned are: tetrabutylammonium iodide, tetrabutylammonium bromide, tributyl-methylphosphonium bromide, trimethyl- $C_{13}/C_{15}$ -alkylammonium chloride, dibenzyl-dimethyl-ammonium methylsulphate, dimethyl- $C_{12}/C_{14}$ -alkyl-benzylammonium

chloride, tetrabutylammonium hydroxide, 15-crown-5, 18-crown-6, triethylbenzylammonium chloride, trimethylbenzylammonium chloride or tris-[2-(2-methoxyethoxy)-ethyl]-amine.

- 5 When carrying out process (b) according to the invention, the reaction temperatures can be varied within a substantial range. In general, the process is carried out at temperatures between  $-20^{\circ}\text{C}$  and  $150^{\circ}\text{C}$ , preferably at temperatures between  $0^{\circ}\text{C}$  and  $20^{\circ}\text{C}$ .
- 10 For carrying out process (b) according to the invention, 1.0 to 2.5 moles, preferably 1.0 to 1.5 moles, of alkylating agent of the formula (V) and, if appropriate, 1.0 to 2.5 moles, preferably 1.0 to 1.5 moles, of base as
- 15 reaction auxiliary and, if appropriate, 0.001 to 2.0 moles, preferably 0.01 to 1.0 mole, of phase transfer catalyst are generally employed per mole of N-aryl-nitrogen heterocycle of the formula (IV).
- 20 The reaction is carried out and the reaction products are worked up and isolated by generally customary methods (cf. in this context the preparation examples).

Suitable ester-cleaving reagents for carrying out the first step of process (c) according to the invention are all customary inorganic and organic acids.

Aqueous mineral acids such as, for example, hydrochloric acid or sulphuric acid, are preferably used.

25

Suitable diluents for carrying out the first step of process (c) according to the invention are all customary organic or inorganic solvents. Solvents which are preferably used are polar, water-miscible organic solvents, in particular alcohols such as methanol, ethanol, propanol or butanol, their mixtures with water, or pure water.

When carrying out the first step of process (c) according to the invention, the reaction temperatures can be varied within a substantial range. In general, the process is carried out at temperatures between  $-20^{\circ}\text{C}$  and  $120^{\circ}\text{C}$ , preferably at temperatures between  $0^{\circ}\text{C}$  and  $80^{\circ}\text{C}$ .

For carrying out the first step of process (c) according to the invention, 1.0 to 10.0 moles, preferably 1.0 to 5.0 moles, of aqueous mineral acids are generally employed per mole of N-aryl-nitrogen heterocycle of the formula (Ia). The reaction is carried out and the reaction products are worked up and isolated by generally known processes.

Suitable diluents for carrying out the second step of process (c) according to the invention are inert organic solvents. These include, in particular, aliphatic, alicyclic or aromatic, optionally halogenated hydrocarbons such as, for example, benzene, toluene, xylene, chlorobenzene, dichlorobenzene, petroleum ether, hexane, cyclohexane, dichloromethane, chloroform, carbon tetrachloride; ethers such as diethyl ether, diisopropyl ether, dioxane, tetrahydrofuran or ethylene glycol

dimethyl ether or ethylene glycol diethyl ether; ketones such as acetone, butanone or methyl isobutyl ketone; nitriles such as acetonitrile, propionitrile or benzonitrile; amides such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide; esters such as methyl acetate or ethyl acetate, or sulfoxides such as dimethyl sulfoxide. It is also possible to use a suitable excess of alcohol of the formula (VII) as reactant and simultaneously as diluent.

If appropriate, the second stage of process (c) according to the invention can be carried out in the presence of a suitable reaction auxiliary. Suitable reaction auxiliaries are all customary inorganic or organic bases. These include, for example, alkaline earth metal hydroxides or alkali metal hydroxides such as sodium hydroxide, calcium hydroxide, potassium hydroxide or else ammonium hydroxide, alkali metal carbonates such as sodium carbonate, potassium carbonate, potassium hydrogen carbonate, sodium hydrogen carbonate or ammonium carbonate, alkali-the alkaline earth metal acetates such as sodium acetate, potassium acetate, calcium acetate or ammonium acetate, and also tertiary amines such as trimethylamine, triethylamine, tributylamine, N,N-dimethylaniline, pyridine, piperidine, N-methylpiperidine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

When carrying out the second step of process (c)

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according to the invention, the reaction temperatures can be varied within a substantial range. In general, the process is carried out at temperatures between  $-20^{\circ}\text{C}$  and  $+150^{\circ}\text{C}$ , preferably at temperatures between  $0^{\circ}\text{C}$  and  $120^{\circ}\text{C}$ .

5 For carrying out the second step of process (c) according to the invention, 1.0 to 10.0 moles, preferably 1.0 to 5.0 moles, of alcohol of the formula (VII) and, if appropriate, 0.001 to 2.0 moles, preferably 0.001 to 1.0 mole, of reaction auxiliary are generally employed per  
10 mole of N-aryl-nitrogen heterocycle of the formula (VI).

The end products of the formula (I) are purified with the aid of customary processes, for example by column chromatography or by recrystallisation.

15 They are characterised with the aid of the melting point or, in the case of non-crystallising compounds, with the aid of proton nuclear resonance spectroscopy ( $^1\text{H}$  NMR).

The active compounds according to the invention can be used as defoliants, desiccants, agents for destroying broad-leaved plants and, especially, as weed-killers. By  
20 weeds, in the broadest sense, there are to be understood all plants which grow in locations where they are undesired. Whether the substances according to the invention act as total or selective herbicides depends essentially on the amount used.

25 The active compounds according to the invention can be used, for example, in connection with the following

plants:

5     Dicotyledon weeds of the genera: Sinapis, Lepidium, Galium, Stellaria, Matricaria, Anthemis, Galinsoga, Chenopodium, Urtica, Senecio, Amaranthus, Portulaca, Xanthium, Convolvulus, Ipomoea, Polygonum, Sesbania, Ambrosia, Cirsium, Carduus, Sonchus, Solanum, Rorippa, Rotala, Lindernia, Lamium, Veronica, Abutilon, Emex, Datura, Viola, Galeopsis, Papaver, Centaurea, Trifolium, Ranunculus and Taraxacum.

10    Dicotyledon cultures of the genera: Gossypium, Glycine, Beta, Daucus, Phaseolus, Pisum, Solanum, Linum, Ipomoea, Vicia, Nicotiana, Lycopersicon, Arachis, Brassica, Lactuca, Cucumis and Cucurbita.

15    Monocotyledon weeds of the genera: Echinochloa, Setaria, Panicum, Digitaria, Phleum, Poa, Festuca, Eleusine, Brachiaria, Lolium, Bromus, Avena, Cyperus, Sorghum, Agropyron, Cynodon, Monochoria, Fimbristylis, Sagittaria, Eleocharis, Scirpus, Paspalum, Ischaemum, Sphenoclea, Dactyloctenium, Agrostis, Alopecurus and Apera.

20    Monocotyledon cultures of the genera: Oryza, Zea, Triticum, Hordeum, Avena, Secale, Sorghum, Panicum, Saccharum, Ananas, Asparagus and Allium.

25    However, the use of the active compounds according to the invention is in no way restricted to these genera, but also extends in the same manner to other plants.

The compounds are suitable, depending on the concentration, for the total combating of weeds, for example on industrial plants and railway lines, and on paths and squares with or without tree plantings.

5 Equally, the compounds can be employed for combating weeds in perennial cultures, for example afforestations, decorative tree plantings, orchards, vineyards, citrus groves, nut orchards, banana plantations, coffee plantations, tea plantations, rubber plantations, oil  
10 palm plantations, cocoa plantations, soft fruit plantings and hopfields, in lawns, turf and pasture-land, and for the selective combating of weeds in annual cultures.

In this context, the active compounds according to the invention can be employed with particularly good success  
15 for combating monocotyledon and dicotyledon weeds in monocotyledon and dicotyledon cultures such as, for example, wheat or cotton.

In addition, the active compounds according to the invention also engage in the metabolism of the plants and  
20 can therefore be employed as growth regulators.

Experience to date of the mode of action of the plant growth regulators has shown that an active compound can also exert several different actions on plants. The actions of the compounds depend essentially on the point  
25 in time at which they are used, relative to the stage of development of the plant, and on the amounts of active compound applied to the plants or their environment and

the way in which the compounds are applied. In each case, growth regulators are intended to influence the crop plants in the particular manner desired.

5 The amount of leaf on plants can be controlled, under the influence of growth regulators, so that defoliation of the plants at a desired point in time is achieved. Such defoliation is of great importance in the mechanical harvesting of cotton, but is also of interest for facilitating harvesting in other crops, such as, for example, 10 viticulture. Defoliation of the plants can also be carried out to lower the transpiration of plants before they are transplanted.

15 The active compounds can be converted into the customary formulations, such as solutions, emulsions, wettable powders, suspensions, powders, dusting agents, pastes, soluble powders, granules, suspension-emulsion concentrates, natural and synthetic materials impregnated with active compound, and very fine capsules in polymeric substances.

20 These formulations are produced in a known manner, for example by mixing the active compounds with extenders, that is liquid solvents and/or solid carriers, optionally with the use of surface-active agents, that is emulsifying agents and/or dispersing agents and/or foam-forming 25 agents.

In the case of the use of water as an extender, organic



solvents can, for example, also be used as auxiliary solvents. As liquid solvents, there are suitable in the main: aromatics, such as xylene, toluene, or alkyl-naphthalenes, chlorinated aromatics and chlorinated  
5 aliphatic hydrocarbons, such as chlorobenzenes, chloro-ethylenes or methylene chloride, aliphatic hydrocarbons, such as cyclohexane or paraffins, for example petroleum fractions, mineral and vegetable oils, alcohols, such as butanol or glycol as well as their ethers and esters,  
10 ketones, such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents, such as dimethylformamide and dimethyl sulphoxide, as well as water.

As solid carriers there are suitable: for example ammonium salts and ground natural minerals, such as kaolins,  
15 clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, and ground synthetic minerals, such as highly disperse silica, alumina and silicates; as solid carriers for granules there are suitable: for  
20 example crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, as well as synthetic granules of inorganic and organic meals, and granules of organic material such as sawdust, coconut shells, maize cobs and tobacco stalks; as emulsifying  
25 and/or foam-forming agents there are suitable: for example non-ionic and anionic emulsifiers, such as polyoxyethylene fatty acid esters, polyoxyethylene fatty alcohol ethers, for example alkylaryl polyglycol ethers, alkylsulphonates, alkyl sulphates, arylsulphonates as

well as albumen hydrolysis products; as dispersing agents there are suitable: for example lignin-sulphite waste liquors and methylcellulose.

5 Adhesives such as carboxymethylcellulose and natural and synthetic polymers in the form of powders, granules or latexes, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, as well as natural phospholipids, such as cephalins and lecithins, and synthetic phospholipids, can be used in the formulations. Further additives can be  
10 mineral and vegetable oils.

It is possible to use colorants such as inorganic pigments, for example iron oxide, titanium oxide and Prussian Blue, and organic dyestuffs, such as alizarin dyestuffs, azo dyestuffs and metal phthalocyanine dyestuffs, and trace nutrients such as salts of iron,  
15 manganese, boron, copper, cobalt, molybdenum and zinc.

The formulations in general contain between 0.1 and 95 per cent by weight of active compound, preferably between 0.5 and 90%.

20 For combating weeds, the active compounds according to the invention, as such or in the form of their formulations, can also be used as mixtures with known herbicides, finished formulations or tank mixes being possible.

25 Suitable herbicides for the mixtures are known herbi-

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5 diflufenican and propanil; arylcarboxylic acids such as,  
for example, dichloropicolinic acid, dicamba and  
picloram; aryloxyalkanoic acids such as, for example, 2,4  
D, 2,4 DB, 2,4 DP, fluroxypyr, MCPA, MCPP and triclopyr;  
10 aryloxy-phenoxy-alkanoic esters such as, for example,  
diclofop-methyl, fenoxaprop-ethyl, fluazifop-butyl,  
haloxyfop-methyl and quizalofop-ethyl; azinones such as,  
for example, chloridazon and norflurazon; carbamates such  
as, for example, chlorpropham, desmedipham, phenmedipham  
15 and propham; chloroacetanilides such as, for example,  
alachlor, acetochlor, butachlor, metazachlor,  
metolachlor, pretilachlor and propachlor; dinitroanilines  
such as, for example, oryzalin, pendimethalin and tri-  
fluralin; diphenyl ethers such as, for example,  
20 acifluorfen, bifenox, fluoroglycofen, fomesafen, halo-  
safen, lactofen and oxyfluorfen; ureas such as, for  
example, chlortoluron, diuron, fluometuron, isoproturon,  
linuron and methabenzthiazuron; hydroxylamines such as,  
for example, alloxydim, clethodim, cycloxydim, sethoxydim  
25 and tralkoxydim; imidazolinones such as, for example,  
imazethapyr, imazamethabenz, imazapyr and imazaquin;  
nitriles such as, for example, bromoxynil, dichlobenil  
and ioxynil; oxyacetamides such as, for example,  
mefenacet; sulphonylureas such as, for example, amido-  
30 sulfuron, bensulfuron-methyl, chlorimuron-ethyl, chlor-  
sulfuron, cinosulfuron, metsulfuron-methyl, nicosulfuron,  
primisulfuron, pyrazosulfuron-ethyl, thifensulfuron-  
methyl, triasulfuron and tribenuron-methyl; thiol-  
carbamates such as, for example, butylate, cycloate, di-  
allate, EPTC, esprocarb, molinate, prosulfocarb, thio-

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5        bencarb and tri-allate; triazines such as, for example,  
         atrazine, cyanazine, simazine, simetryne, terbutryne and  
         terbutylazine; triazinones such as, for example,  
         hexazinone, met amitron and metribuzin; others such as,  
         for example, aminotriazole, benfuresate, bentazone,  
         cinmethylin, clomazone, clopyralid, difenzoquat,  
         dithiopyr, ethofumesate, fluorochloridone, glufosinate,  
10        glyphosate, isoxaben, pyridate, quinchlorac, quinmerac,  
         sulphosate and tridiphane.

15        Mixtures with other known active compounds, such as  
         fungicides, insecticides, acaricides, nematocides, bird  
         repellants, plant nutrients and agents which improve soil  
         structure, are also possible.

20        The active compounds can be used as such, in the form of  
         their formulations or in the use forms prepared therefrom  
         by further dilution, such as ready-to-use solutions,  
         suspensions, emulsions, powders, pastes and granules.

25

30

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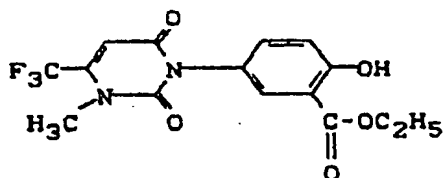
They are used in the customary manner, for example by watering, spraying, atomising or scattering.

The active compounds according to the invention can be applied either before or after emergence of the plants.

- 5 They can also be incorporated into the soil before sowing.

- 10 The amount of active compound used can vary within a substantial range. It depends essentially on the nature of the desired effect. In general, the amounts used are between 1 g and 10 kg of active compound per hectare of soil surface, preferably between 5 g and 5 kg per ha.

The preparation and use of the active compounds according to the invention can be seen from the following examples.

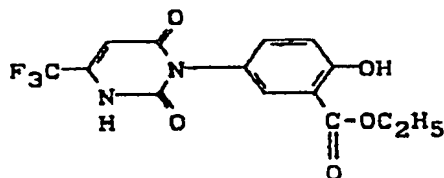
Preparation Examples:Example 1:

(Process (b))

0.8 g (0.006 mol) of anhydrous potassium carbonate and  
 5 1 g (0.006 mol) of methyl iodide are added in succession  
 to 2.1 g (0.006 mol) of 3-(4-hydroxy-3-ethoxycarbonyl-  
 phenyl)-6-trifluoromethyl-1,2,3,4-tetrahydro-1H-pyrimi-  
 dine-2,4-dione in 50 ml of dimethyl sulphoxide, and the  
 mixture is stirred for 16 hours at room temperature. For  
 10 working-up, the reaction mixture is poured into water,  
 acidified using hydrochloric acid and extracted using  
 dichloromethane. The organic phase is dried over magnes-  
 ium sulphate and concentrated in vacuo, and the residue  
 is purified by chromatography on silica gel (eluent:  
 15 dichloromethane).

1 g (46.5% of theory) of 3-(4-hydroxy-3-ethoxycarbonyl-  
 phenyl)-1-methyl-6-trifluoromethyl-1,2,3,4-tetrahydro-  
 pyrimidine-2,4-dione of melting point 65°C is obtained.

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Preparation of the starting compoundExample IV-1:

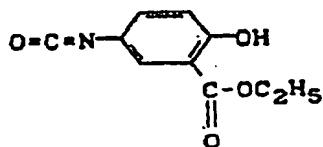
## (Process (a))

5 9.15 g (0.05 mol) of ethyl 3-amino-4,4,4-trifluoro-  
 crotonate (cf., for example, J. Heterocycl. Chem. 9,513  
 [1972]) in 20 ml of dimethylformamide and 10.35 g  
 (0.05 mol) of 4-hydroxy-3-ethoxycarbonyl-phenyl isocyan-  
 ate in 100 ml of toluene are added dropwise with stirring  
 at -30°C to a suspension, prepared at 0°C, of 3.0 g  
 10 (0.1 mol) of sodium hydride (80% in paraffin oil) in  
 50 ml of dimethylformamide, and, when the addition has  
 ended, the mixture is stirred for 6 hours at -30°C. For  
 working-up, the reaction mixture is poured into water,  
 acidified using hydrochloric acid and extracted using  
 15 ethyl acetate. The organic phase is dried over magnesium  
 sulphate and freed from solvent in vacuo.

10.3 g (60% of theory) of 3-(4-hydroxy-3-ethoxycarbonyl-  
 phenyl)-6-trifluoromethyl-1,2,3,4-tetrahydro-1H-pyrimi-  
 dine-2,4-dione of melting point 157-59°C are obtained.

Example II-1:

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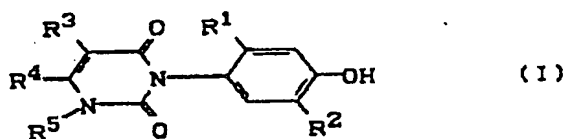


5 9.9 g (0.05 mol) of trichloromethyl chloroformate are added dropwise with stirring at 70°C to 9.05 g (0.05 mol) of ethyl 5-amino-2-hydroxybenzoate (cf., for example, EP 291,159) in 150 ml of ethyl acetate, and the mixture is refluxed for two hours. For working-up, the reaction mixture is concentrated in vacuo, and the residue is taken up in 100 ml of anhydrous toluene.

10 The resulting solution of 4-hydroxy-3-ethoxycarbonyl-phenyl isocyanate is reacted further directly without isolation.

IR (NaCl):  $\nu = 2272 \text{ cm}^{-1}$

15 The following N-aryl-nitrogen heterocycles of the general formula (I) are obtained in an analogous manner and following the general preparation instructions:





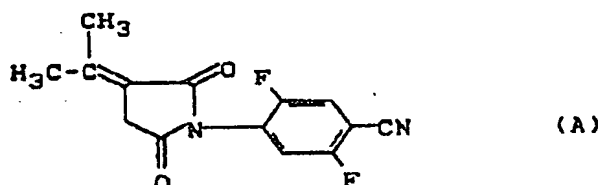
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Ex. No.	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	Physical properties
2	H	-C(O)-O-iC <sub>3</sub> H <sub>7</sub>	H	CF <sub>3</sub>	CH <sub>3</sub>	J=3.39 (s; 3H)
3	H	-C(O)OCH <sub>3</sub>	H	CF <sub>3</sub>	CH <sub>3</sub>	m.p. 185°C
4	F	-C(O)-O-iC <sub>3</sub> H <sub>7</sub>	H	CF <sub>3</sub>	CH <sub>3</sub>	?
IV-2	H	-C(O)-O-CH <sub>3</sub>	H	CF <sub>3</sub>	H	m.p. 201°C

The <sup>1</sup>H-NMR spectra were recorded in deuteriochloroform (CDCl<sub>3</sub>) with tetramethylsilane (TMS) as the internal standard. The data given are the chemical shift as δ value in ppm.

Use Examples:

In the use examples which follow, the compound listed below was employed as comparison substance:



1-(4-Cyano-2,5-difluorophenyl)-3-(1-methylethylidene)-pyrrolidine-2,5-dione  
(cf., for example, DE 3,835,168)

Example A:Post-emergence test

Solvent: 5 parts by weight of acetone

Emulsifier: 1 part by weight of alkylaryl polyglycol  
ether

5

To produce a suitable preparation of active compound,  
1 part by weight of active compound is mixed with the  
stated amount of solvent, the stated amount of emulsifier  
is added, and the concentrate is diluted with water to  
the desired concentration.

10

Test plants which have a height of 5 to 15 cm are sprayed  
with the preparation of active compound in such a way  
that the amounts of active compound desired in each case  
are applied per unit area. The concentration of the spray  
liquor is chosen in such a way that the amounts of active  
compound desired in each case are applied in 2,000 l of  
water per hectare. After three weeks, the degree of  
damage to the plants is rated in % damage compared with  
the development of the untreated control.

15

The figures denote:

20

0% = no action (like untreated control)  
100% = total destruction

A clearly superior activity combined with an equally good  
crop plant selectivity compared with the prior art is

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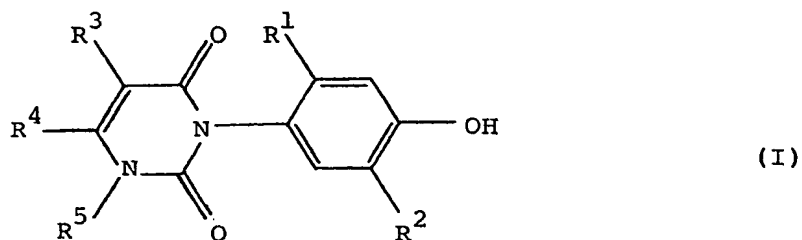
shown, in this test, for example by the compound of Preparation Example 1.

It will be understood that the specification and examples are illustrative but not limitative of the present invention and that other embodiments within the spirit and scope of the invention will suggest themselves to those skilled in the art.

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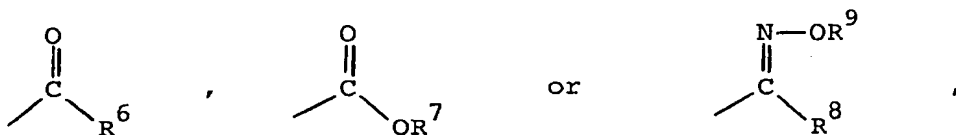
THE EMBODIMENTS OF THE INVENTION IN WHICH AN EXCLUSIVE  
PROPERTY OR PRIVILEGE IS CLAIMED ARE DEFINED AS FOLLOWS:

1. A N-aryl-nitrogen heterocycle of the general formula  
(I)



wherein

$R^1$  represents hydrogen or halogen,  
 $R^2$  represents one of the radicals



$R^3$  represents hydrogen and  
 $R^4$  represents alkyl or halogenoalkyl, or  
 $R^3$  and  $R^4$  together represent a divalent alkanediyl  
radical and

$R^5$  represents in each case straight-chain or branched  
alkyl, alkenyl, alkynyl, halogenoalkyl or cycloalkyl,  
where

$R^6$  represents hydrogen or alkyl,  
 $R^7$  represents in each case optionally substituted  
alkyl, alkenyl or cycloalkyl,

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$R^8$  represents hydrogen or alkyl and

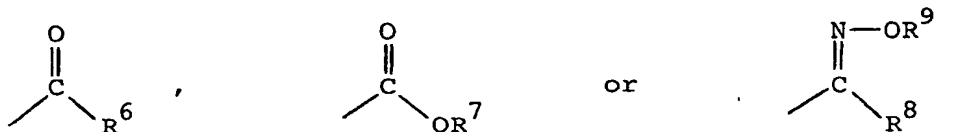
$R^9$  represents in each case optionally substituted alkyl, alkenyl or cycloalkyl.

2. A N-aryl-nitrogen heterocycle of the general formula (I) according to claim 1,

wherein

$R^1$  represents hydrogen, fluorine, chlorine, bromine or iodine,

$R^2$  represents one of the radicals



$R^3$  represents hydrogen and

$R^4$  represents in each case straight-chain or branched alkyl having 1 to 6 carbon atoms or halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, or

$R^3$  and  $R^4$  together represent a divalent straight-chain or branched alkanediyl radical having 3 to 10 carbon atoms, and

$R^5$  represents in each case straight-chain or branched alkyl having 1 to 6 carbon atoms, halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, alkenyl having 2 to 6 carbon atoms, alkynyl having 2 to 6 carbon atoms, or cycloalkyl having 3 to 8 carbon atoms, where

$R^6$  represents hydrogen or straight-chain or branched alkyl having 1 to 6 carbon atoms,

$R^7$  represents in each case straight-chain or branched alkyl having 1 to 12 carbon atoms, halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, alkoxyalkyl, alkoxyalkoxyalkyl or alkoxycarbonylalkyl, each of which has 1 to 6 carbon atoms in the individual alkyl moieties, alkenyl having 2 to 6 carbon atoms, or cycloalkyl which has 3 to 8 carbon atoms and which is optionally monosubstituted or polysubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 4 carbon atoms,

$R^8$  represents hydrogen or straight-chain or branched alkyl having 1 to 6 carbon atoms and

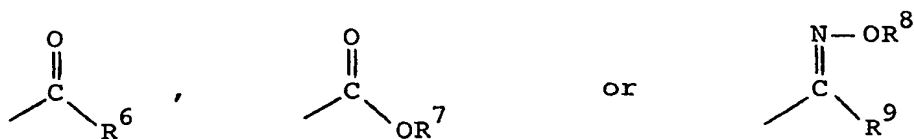
$R^9$  represents in each case straight-chain or branched alkyl having 1 to 12 carbon atoms, halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, alkoxyalkyl, alkoxyalkoxyalkyl or alkoxycarbonylalkyl, each of which has 1 to 6 carbon atoms in the individual alkyl moieties, alkenyl having 2 to 6 carbon atoms, or cycloalkyl which has 3 to 8 carbon atoms and which is optionally monosubstituted or polysubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 4 carbon atoms.

3. A N-aryl-nitrogen heterocycle of the general formula (I) according to claim 1, wherein

$R^1$  represents hydrogen, fluorine, chlorine or bromine,

$R^2$  represents one of the radicals

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$\text{R}^3$  represents hydrogen and

$\text{R}^4$  represents in each case straight-chain or branched alkyl having 1 to 4 carbon atoms or halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, or

$\text{R}^3$  and  $\text{R}^4$  together represent a divalent straight-chain or branched alkanediyl radical having 3 to 8 carbon atoms, and

$\text{R}^5$  represents in each case straight-chain or branched alkyl having 1 to 4 carbon atoms, halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, alkenyl having 2 to 5 carbon atoms, alkynyl having 2 to 5 carbon atoms, or cycloalkyl having 3 to 7 carbon atoms,

where

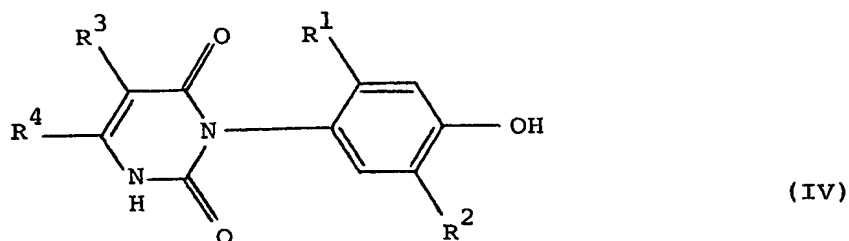
$\text{R}^6$  represents hydrogen or straight-chain or branched alkyl having 1 to 3 carbon atoms,

$\text{R}^7$  represents in each case straight-chain or branched alkyl having 1 to 8 carbon atoms, halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, alkoxyalkyl, alkoxyalkoxyalkyl or alkoxycarbonylalkyl, each of which has 1 to 4 carbon atoms in the individual alkyl moieties, alkenyl having 2 to 4 carbon atoms, or cycloalkyl which has 3 to 7 carbon atoms and is optionally monosubstituted to trisubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 3 carbon atoms,

$R^8$  represents hydrogen or straight-chain or branched alkyl having 1 to 3 carbon atoms and

$R^9$  represents in each case straight-chain or branched alkyl having 1 to 8 carbon atoms, halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, alkoxyalkyl, alkoxyalkoxyalkyl or alkoxycarbonylalkyl, each of which has 1 to 4 carbon atoms in the individual alkyl moieties, alkenyl having 2 to 4 carbon atoms, or cycloalkyl which has 3 to 7 carbon atoms and is optionally monosubstituted to trisubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 3 carbon atoms.

4. A N-aryl-nitrogen heterocycle of the general formula (IV)

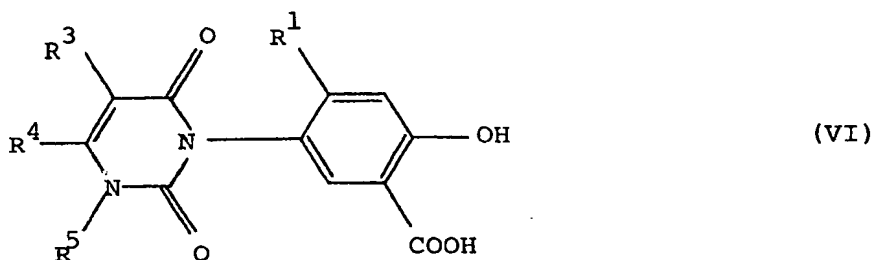


wherein  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are as defined in claim 1.

5. A N-aryl-nitrogen heterocycle of the general formula (VI)

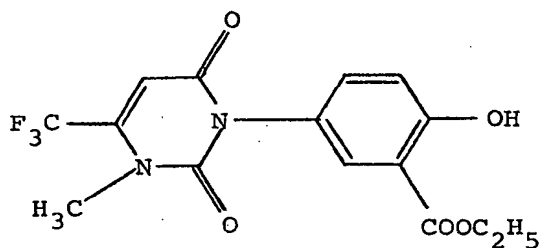


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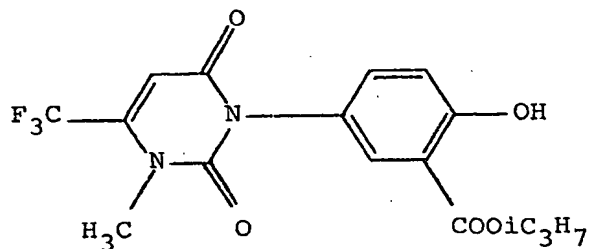


wherein  $R^1$ ,  $R^3$ ,  $R^4$  and  $R^5$  are as defined in claim 1.

6. 3-(4-Hydroxy-3-ethoxycarbonylphenyl)-1-methyl-6-trifluoromethyl-1,2,3,4-tetrahydropyrimidine-2,4-dione of the formula

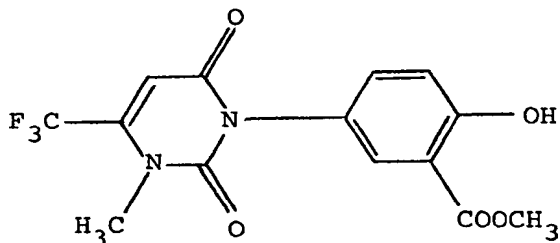


7. 3-(4-Hydroxy-3-isopropoxycarbonylphenyl)-1-methyl-6-trifluoromethyl-1,2,3,4-tetrahydropyrimidine-2,4-dione of the formula

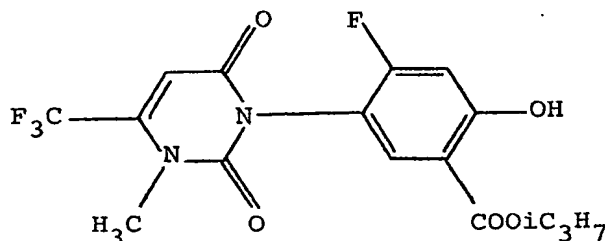


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8. 3-(4-Hydroxy-3-methoxycarbonylphenyl)-1-methyl-6-trifluoromethyl-1,2,3,4-tetrahydropyrimidine-2,4-dione of the formula



9. 3-(6-Fluoro-4-hydroxy-3-isopropoxy carbonylphenyl)-1-methyl-6-trifluoromethyl-1,2,3,4-tetrahydropyrimidine-2,4-dione of the formula



10. A herbicidal composition comprising a herbicidally effective amount of a compound according to any one of claims 1 to 3 and 6 to 9 in admixture with a suitable carrier or diluent.

11. A herbicidal composition comprising a herbicidally effective amount of a compound according to any one of claims 1 to 3 and 6 to 9 in admixture with a solid diluent or carrier, a liquified normally gaseous diluent or carrier, or a liquid

diluent or carrier containing a surface active agent.

12. A method of combating weeds which comprises applying to the weeds, or to a habitat thereof, a herbicidally effective amount of a compound according to any one of claims 1 to 3 and 6 to 9.

13. A method of combating weeds which comprises applying to the weeds, or to a habitat thereof, a herbicidally effective amount of a composition containing a compound according to any one of claims 1 to 3 and 6 to 9 in admixture with a suitable carrier or diluent.

14. A method of combating weeds which comprises applying to the weeds, or to a habitat thereof, a herbicidally effective amount of a composition containing between 0.1 and 95 % by weight of a compound according to any one of claims 1 to 3 and 6 to 9 in admixture with a suitable carrier or diluent.

15. A method of combating weeds which comprises applying to the weeds, or to a habitat thereof, a herbicidally effective amount of a composition containing between 0.5 to 90 % by weight of a compound according to any one of claims 1 to 3 and 6 to 9 in admixture with a suitable carrier or diluent.

16. A method of combating weeds which comprises applying to the weeds, or to a habitat thereof, a herbicidally effective amount of a compound according to any one of claims 1 to 3 and 6 to 9 wherein the compound is applied as a pre-emergence herbicide.

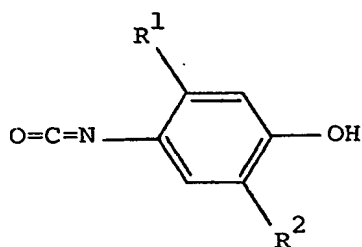
17. A method of combating weeds which comprises applying to the weeds, or to a habitat thereof, a herbicidally effective amount of a compound according to any one of claims 1 to 3 and 6 to 9 wherein the compound is applied as a post-emergence herbicide.

18. A method of combating weeds which comprises applying to the weeds, or to a habitat thereof, a herbicidally effective amount of a compound according to any one of claims 1 to 3 and 6 to 9 wherein the compound is applied to an area of cultivation at a rate of between 1 g and 10 kg/ha.

19. A method of combating weeds which comprises applying to the weeds, or to a habitat thereof, a herbicidally effective amount of a compound according to any one of claims 1 to 3 and 6 to 9 wherein the compound is applied to an area of cultivation at a rate of between 5 g and 5 kg/ha.

20. A process for preparing a compound of formula (I) as defined in claim 1, wherein  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$  and  $R^5$  are as defined in claim 1, which process comprises

a) reacting an isocyanate of the formula (II)

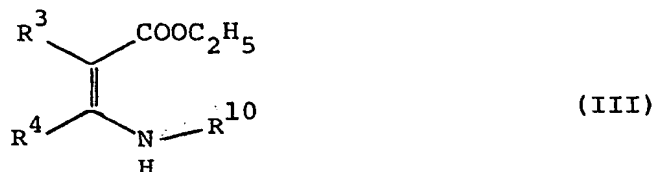


(II)

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in which

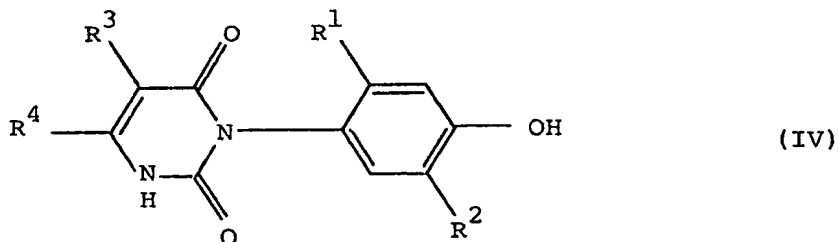
$R^1$  and  $R^2$  have the above-mentioned meanings, with [a]  $\beta$ -enamino ester of the formula (III)



in which

$R^3$  and  $R^4$  have the above-mentioned meanings and  $R^{10}$  represents hydrogen or in each case straight-chain or branched alkyl, alkenyl, alkynyl, halogenoalkyl or cycloalkyl, and, where required

b) reacting the resulting N-aryl-nitrogen heterocycle of the formula (IV)



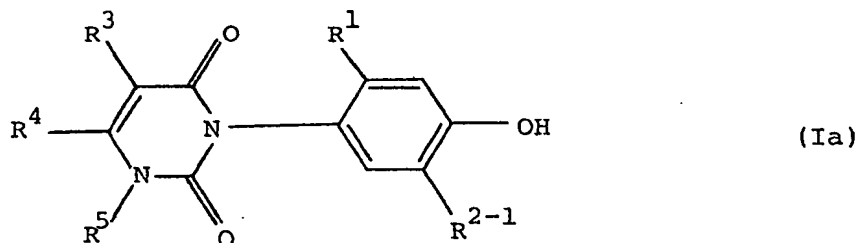
in which

$R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  have the above-mentioned meanings with an alkylating agent of the formula (V)



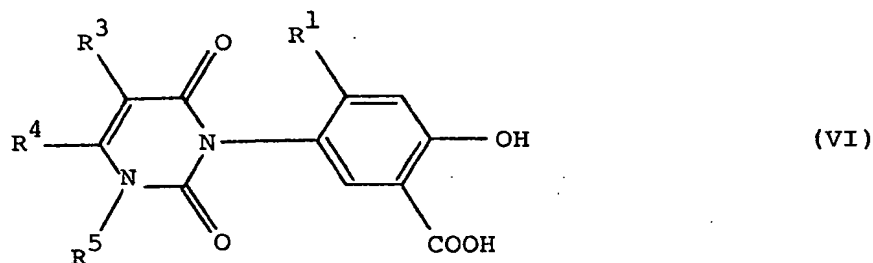
in which

$R^5$  has the above-mentioned meaning and  
 E represents an electron-attracting leaving group, or by  
 c) first hydrolysing an N-aryl-nitrogen heterocycle  
 obtained with the aid of process (a) or (b) of the formula (Ia)



in which

$R^1$ ,  $R^3$ ,  $R^4$  and  $R^5$  have the above-mentioned meanings and  
 $R^{2-1}$  represents an alkoxycarbonyl radical, with an acid  
 on the ester group in the aryl moiety, and esterifying the  
 resulting N-aryl-nitrogen heterocycle of the formula (VI)



in which

$R^1$ ,  $R^3$ ,  $R^4$  and  $R^5$  have the above-mentioned meanings,  
 in a subsequent 2nd step with an alcohol of the formula (VII)



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in which

R<sup>7</sup> has the above-mentioned meaning.

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OTTAWA, CANADA

PATENT AGENTS

**SUBSTITUTE**

***REMPLACEMENT***

**SECTION is not Present**

***Cette Section est Absente***